Tumbling of polymers in random flow with mean shear

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We consider tumbling of a polymer molecule in a permanent shear flow. Tumbling is driven by either random velocity or thermal fluctuations. We find that: The probability distribution of the polymer's orientation has a maximum near the direction determined by the shear but it also shows long, algebraic, tails. The probability distribution of the time between two subsequent flips of the polymer is characterized by an exponential tail.

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With the development of novel optical methods, a number of high quality experimental observations focusing on resolving dynamics of individual polymers (DNA molecules) subjected to a non-homogeneous flow have been reported [1–4]. One noticeable phenomenon observed in shear flows (with the shear rate, s, being much larger than the inverse polymer relaxation time) is the effect of tumbling: The molecule which spends most of the time being oriented along the major direction of the shear sometimes and suddenly (aperiodically) makes a flip in the direction dictated by the shear. The aperiodic nature of the tumbling turns, provoked by the thermal fluctuations (Langevin forces), was related to a broad (algebraic like) form of the power spectral density per unit time of the extension fluctuations. The dependence of the typical tumbling time, where the tumbling time is defined as the time between two subsequent flips of the polymer, on the shear rate was measured [3] and subsequently confirmed in theoretical/numerical analysis [5].

In another experimental breakthrough, Groisman and Steinberg have discovered that a chaotic flow state, called by the authors "elastic turbulence", is observed in dilute and highly viscous polymer solutions [6, 7]. This elastic turbulence flow consists of regular (shear-like) and chaotic components, the latter being weak in comparison with the former one. Resolving an individual polymer in this chaotic state was more challenging than in the regular flow experiment of Refs. [3, 4] but it was still an accessible task [8]. Thus, the coil-stretch transition, predicted by Lumley [9] (see also Ref. [10]) for a chaotic flow, was for the first time observed in direct single-polymer measurements [8]. The tumbling phenomenon was also observed in the elastic turbulence experimental setting [8], but it was not yet explored in detail.

This letter presents a theoretical analysis of the tumbling phenomenon in a chaotic flow with a large mean shear, e.g. of the type correspondent to the elastic turbulence experiments [6–8]. The overall effect of the flow is assumed to be strong enough so that the polymer is strongly stretched. The stretched state is formed as a result of a balance between the stretching (related to spatial inhomogeneity of the flow) and the elasticity (related to the internal molecule relaxation) above the coil-stretched transition [11] when the effect of the thermal noise on the polymer dynamics is negligible. We predict that in this setting, very much like in the case of a permanent shear and thermal fluctuations, the polymer experiences random (aperiodic) tumbling. Our study is focused on, first, establishing the universal features of the statistics of the polymer orientation and, second, on analyzing the statistics of the polymer's tumbling time. We demonstrate that the main results of our analysis are also applicable to tumbling driven by thermal fluctuations.

This letter is organized as follows. We begin by introducing the basic dumb-bell equation governing dynamics of the polymer end-to-end vector in a non-homogeneous flow, and showing that in the stretched state (independent of the type of flow) the angular/orientational part of the polymer dynamics decouples from its extensional counterpart. We examine the orientational statistics of a stretched polymer in a random statistically stationary flow with a mean shear which is much stronger than the velocity fluctuations. Our major results are formulated for the general setting, and then they are illustrated and confirmed for a model of Gaussian and short-time-correlated fluctuations of velocity. The orientation of the polymer is parameterized in terms of the two angles: ϕ, the angle between the projection on the shear plane of the the polymer end-to-end vector and the direction of the shear component of velocity, and θ, measuring the angle between the end-to-end vector and the shear plane (see Fig. 1). We show that due to the weakness of the velocity fluctuations the polymer tends to align with the direction of shear, i.e. typical values of the angles are small. The Probability Distribution Function (PDF) of the angle ϕ is studied first. We find that for ϕ larger than its typical value, ϕ_t, the PDF of ϕ decays as $\mathcal{P}(\phi) \propto 1/\sin^2\phi$. Next, we describe the statistics of the tumbling time. We show that since the tumbling is driven by the weakly fluctuating part of the velocity, the typical tumbling time, τ_t, is large compared to the inverse shear rate, s^{-1}. More precisely, $\tau_t \sim (s\phi_t)^{-1}$. We also find that the PDF of τ has a maximum at $\tau \sim \tau_t$ and the width of the distribution around the maximum is of the order of the same τ_t. The

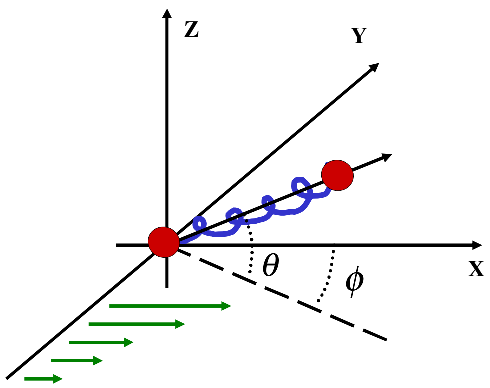

FIG. 1: Schematic figure explaining polymer orientation geometry.

PDF shows an extended (exponential) tail for $\tau \gg \tau_t$. Finally, the statistics of the off-shear plane angular dynamics, controlled by θ, is discussed. One finds that the PDF decays algebraically in the domain of untypically large angles, $1 \gg \theta \gg \phi_t$. The correlation time of θ is also approximately τ_t. Finally, we show that the results formulated above extend to the case of tumbling driven by thermal fluctuations. Some general discussion of the analysis applicability and relevance to real experimental setting concludes the letter.

Polymer extension can be characterized by its end-to-end separation vector, $\boldsymbol{R}$, satisfying the following equation [12, 13]:

$$\partial_t R_i = R_j \nabla_j v_i - \gamma R_i + \zeta_i \,, \tag{1}$$

where the relaxation rate γ is a function of the absolute value R of the vector $\boldsymbol{R}$, the velocity gradient $\nabla_j v_i$ is taken at the molecule position, and ζ_i is the Langevin force. The velocity difference between the polymer end points is approximated in Eq. (1) by the first term of its Taylor expansion in the end-to-end vector. For the elastic turbulence this approximation is justified because the velocity fluctuation spectrum decays in the Fourier space sufficiently fast and thus the relevant part of the velocity is large-scale [6].

Noticeably, if the Langevin force in Eq. (1) can be neglected, then the polymer angular (orientational) dynamics described by the unit vector $\boldsymbol{n} = \boldsymbol{R}/R$ decouples from the dynamics of the end-to-end polymer length:

$$\partial_t n_i = n_j(\delta_{ik} - n_i n_l)\nabla_j v_l \,. \tag{2}$$

The relaxation term (proportional to γ) in Eq. (1) does not enter Eq. (2), thus making the polymer orientational dynamics insensitive to the dynamics/statistics of the polymer extension. Let us choose the coordinate frame as shown in Fig. 1. The mean flow is characterized by the velocity $(sy, 0, 0)$ and the polymer angular dynamics is conveniently parameterized in terms of the angles ϕ and θ: $n_x = \cos\theta\cos\phi$, $n_y = \cos\theta\sin\phi$, $n_z = \sin\theta$. Then Eq. (2) becomes

$$\partial_t \phi = -s\sin^2\phi + \xi_\phi \,, \tag{3}$$

$$\partial_t \theta = -s\frac{\sin(2\phi)}{2}\sin\theta + \xi_\theta \,, \tag{4}$$

where ξ_ϕ and ξ_θ are zero-mean random variables related to the chaotic contribution to the velocity gradient. The statistics of both ξ_ϕ and ξ_θ are assumed to be homogeneous in time.

If the random component of the flow is weak, the respective chaotic terms on the right-hand sides of Eqs. (3,4) are relevant for small ϕ and θ, i.e. only where the regular shear terms are small. Thus, one identifies two distinct regimes: a stochastic one, $|\phi| \lesssim \phi_t$, where ϕ wanders randomly in time, and deterministic one, where the random contribution, ξ_ϕ, can be neglected and ϕ decreases uniformly with time. In accordance with the definition, ϕ_t sets an angular scale where the mean shear and random components are of the same order, $\phi_t \ll 1$ due to the assumed weakness of the velocity fluctuations. Comparing the left-hand side of Eq. (3) with the regular term from its right-hand side (which can be approximated by $s\phi^2$ at $|\phi| \ll 1$) one obtains a relation between the amplitude ϕ_t and the duration τ_t of the stochastic stage: $\tau_t = (s\phi_t)^{-1}$. Thus $\tau_t \gg s^{-1}$, i.e. the stochastic evolution is slower than the deterministic one. Taking into account the last term in Eq. (3), one estimates $\int_0^{\tau_t} dt\, \xi_\phi \sim \phi_t$.

Let us consider the PDF of ϕ, $\mathcal{P}(\phi)$, which is a periodic function of ϕ with the period π, thus it is sufficient to consider only a half-circle. We choose $-\pi/2 < \phi < \pi/2$. The PDF relaxes with the characteristic time τ_t to a stationary distribution. The stationary PDF has a maximum at a positive angle, $\phi \sim \phi_t$, and the PDF's width is of the order of the same ϕ_t. The PDF's tail corresponding to the deterministic region outside the stochastic ϕ_t-narrow domain, $|\phi| \gg \phi_t$, is governed by the Fokker-Planck equation, $\partial_t \mathcal{P} = s\partial_\phi(\sin^2\phi\mathcal{P})$, that contains no "diffusive", i.e. second-order terms in ∂_ϕ. The equation has an obvious stationary solution $\mathcal{P}(\phi) = c\sin^{-2}(\phi)$, characterized by a nonzero average "angular flux", $\langle\partial_t\phi\rangle = -\pi sc$, corresponding to the preferred (clockwise) direction of tumbling. The normalization condition $\int d\phi\,\mathcal{P}(\phi) = 1$, fixes the constant c. Since the major contribution to the normalization integral comes from the region $\phi \sim \phi_t$, where $\mathcal{P} \sim c\phi_t^{-2}$, one estimates $c \sim \phi_t$.

The strong separation of the temporal scales, correspondent to the fast regular rotation, $\sim 1/s$, and to the slow stochastic evolution, $\sim \tau_t$, means that the overall tumbling time, τ, separating two subsequent flips of the polymer is well defined. The major contribution to τ originates from long stochastic wandering in a ϕ_t-narrow vicinity of $\phi = 0$. Therefore, the position of the maximum in the PDF of τ and the width of the peak are both estimated by τ_t. Being interested in the PDF tail, corresponding to $\tau \gg \tau_t$, one observes that if a flip does not occur for a long time, then the delay can be interpreted in terms of a large number, $N = \tau/\tau_t$, of independent unsuccessful attempts to pass (clock-wise in ϕ) the stochastic domain. The probability P of the delayed tumbling event is given by the product of the probabilities of N events, that is $\ln P \sim -\tau/\tau_t$, resulting in the exponential tail of the PDF of τ at $\tau \gg \tau_t$.

To illustrate the general picture explained above, we examine the model in which the velocity fluctuations are short-time correlated and possess Gaussian statistics. Then the statistics of the random variable ξ_ϕ in Eq. (3) is completely determined by the pair correlation function

$$\langle \xi_\phi(t)\xi_\phi(t')\rangle = D\,\delta(t-t')\,. \tag{5}$$

Generally speaking, the coefficient D in Eq. (5) is a function of the angles ϕ and θ. However, noticing that the random variables in Eqs. (3,4) are relevant only at small ϕ and θ, one assumes $D = \text{const}$. Then the statistics of ϕ decouples, i.e. it becomes independent of θ. The weakness of the velocity fluctuations means $D \ll s$. One obtains from Eqs. (3,5) the following Fokker-Planck equation for the PDF of ϕ:

$$\left(\partial_t - s\partial_\phi \sin^2\phi - \frac{D}{2}\partial_\phi^2\right)\mathcal{P}(t,\phi) = 0\,. \tag{6}$$

The stationary solution of Eq.(6) is

$$\mathcal{P} \propto \int^{\phi} d\varphi\, \exp\left\{\frac{s}{D}\left[\phi - \varphi + \frac{\sin(2\varphi)-\sin(2\phi)}{2}\right]\right\}. \tag{7}$$

The integration and normalization constants in (7) should be chosen in such a way that periodicity of $\mathcal{P}$ and the probability normalization conditions are enforced. Eq. (7) shows that ϕ reaches a maximum at $\phi \sim \phi_t$, where $\phi_t = (D/s)^{1/3}$, that estimates the size of the stochastic region in the δ-correlated model. The width of the "hat"-portion of the distribution is estimated by the same ϕ_t, while for $|\phi| \gg \phi_t$ the PDF behaves $\propto \sin^{-2}\phi$, in accordance with the previous general analysis.

To study the PDF of the tumbling time one can use the same Fokker-Planck equation (6), however taking a special (non-periodic) solution corresponding to zero probability flux through the point $\phi = \pi/2$, while the probability flow through the point $\phi = -\pi/2$ is allowed. These conditions guarantee that multiple flips (i.e. clockwise re-entrances of ϕ at $-\pi/2$) are not accounted for, i.e. if ϕ crossed $\pi/2$ in the past the event is not taken into account in the probability. The PDF $\mathcal{P}(t,\phi)$, defined this way, measures a probability that within the observation time t the polymer orientation angle ϕ never crossed $-\pi/2$. Then the total probability for ϕ not to make a single flip within the time τ is given by $\int d\phi\,\mathcal{P}(\tau,\phi)$, which obviously decreases with τ due to the probability flux through $\phi = -\pi/2$. Asymptotically for $t \gg \tau_t$ the corresponding solution is $\mathcal{P}(t,\phi) \to \exp(-E_0 t)\mathcal{P}_1(\phi)$, where E_0 is the smallest eigenvalue of the differential operator in Eq. (6), and $\mathcal{P}_1(\phi)$ is the corresponding eigenfunction which is concentrated at $\phi \sim \phi_t$. At large positive values of ϕ, $\phi \gg \phi_t$, $\ln\mathcal{P}_1 \sim -(s/D)[\phi - \sin(2\phi)/2]$, and for negative ϕ, $|\phi| \gg \phi_t$, one recovers $\mathcal{P}_1 \to c_1 \sin^{-2}\phi$. This gives the behavior correspondent to a non-zero probability flux towards negative ϕ. Similar to the stationary case, one obtains $c_1 \sim \phi_t$. Integrating Eq. (6) over ϕ, one derives $sc_1 = E_0\int d\phi\,\mathcal{P}_1$. Since $\int d\phi\,\mathcal{P}_1 \sim 1$, one estimates $E_0 \sim 1/\tau_t$. Thus one arrives at the exponential behavior discussed previously.

Next, we describe the statistics of θ. Being interested in the domain $\theta \ll 1$, one approximates, $\sin\theta \approx \theta$, thus deriving from Eq. (4)

$$\theta(0) = \int_{-\infty}^{0} dt\, \exp[\rho(t)]\xi_\theta(t) \tag{8}$$

$$\partial_t\rho = (s/2)\sin(2\phi)\,, \quad \rho(0) = 0\,. \tag{9}$$

In spite of the fact that for the regular, $\phi \sim 1$, part of the dynamics one estimates, $\partial_t\rho \sim s$, the polymer spends most of time at $\phi \ll 1$. Thus typically $\partial_t\rho \approx s\phi$, which results in, $\langle\partial_t\rho\rangle \sim s\phi_t \sim 1/\tau_t$. (Notice, that the average $\langle\partial_t\rho\rangle$ measures the Lyapunov exponent, λ, of the flow, which is thus estimated as $\lambda \sim 1/\tau_t$.) Fluctuations of $\partial_t\rho$ are correlated over times of the order of τ_t. Since $\langle\partial_t\rho\rangle > 0$, the typical ρ is an increasing function of time t which becomes of order unity in its absolute value within $|t| \sim \tau_t$. Therefore, the integral on the right-hand side of Eq. (8) is formed at $t \sim \tau_t$ and one concludes that the correlation time of θ is given by the same τ_t. Thus, the characteristic value of θ can be estimated as $\int_0^{\tau_t} dt\,\xi_\theta$, in complete analogy with ϕ. Assuming $\xi_\theta \sim \xi_\phi$, one finds that the characteristic value of θ is estimated by the same quantity ϕ_t.

Our next task is to examine the tail of the PDF of θ corresponding to $|\theta| \gg \phi_t$. The main contribution to the tail comes from rare fluctuations of a special kind: ρ starts from $-\infty$ at $t = -\infty$ reaches a maximum value $\rho_0 > 0$ at some negative time, $t = -T$ ($T \gg \tau_t$), and decreases later becoming zero at $t = 0$. Then the main contribution to the integral (8) originates from times surrounding $-T$ and one derives, $\theta = \exp(\rho_0)\tilde{\xi}_\theta$, where $\tilde{\xi}_\theta$ is an integral of ξ_θ taken over a τ_t-wide vicinity of $t = -T$. Thus according to the large deviation theory [14] the PDF of ρ_0 (which is an integral of a random process over the interval $[-T, 0]$) can be written as $\sim \exp[-(T/\tau_t)f(\rho_0\tau_t/T)]$, where $f(x)$ is a dimensionless function, dependent on details of the ϕ statistics. Next, the statistics of $\tilde{\xi}_\theta$ is stationary, and the typical $\tilde{\xi}_\theta$ is determined ϕ_t, thus the PDF of $\tilde{\xi}_\theta$ can be presented as $\exp[-g(\tilde{\xi}_\theta/\phi_t)]$, where $g(x)$ is a dimensionless function. Combining these observations, one obtains the following expression for the probability of the event: $\exp[-g(\theta\exp[-\rho_0]/\phi_t)] - f(\tau_t\rho_0/T)T/\tau_t]$, which still needs to be optimized (integrated) with respect to both T and ρ_0. The result of the saddle-point evaluation is $\rho_0 \approx \ln(\theta/\phi_t)$, $T = \rho_0 x\tau_t$, where x solves $f(x) = xf'(x)$. Finally, one arrives at $\mathcal{P}(\theta) \propto |\theta|^{-f'(x)}$, which is valid for $1 \gg |\theta| \gg \phi_t$. Estimating $f(x)$ is not simple, but it is obvious that $f'(x) = O(1)$.

Let us briefly discuss the relation of our results to the behavior of polymers in permanent shear flow and subjected to thermal fluctuations. One finds that the phenomenon of tumbling generated by thermal fluctuations is very much similar to the one generated by random fluctuations of velocity. This is due to the universal validity of the following major observation: The fluctuations are essential only in a narrow angular strip where the polymer spends most of the time. Therefore, recalling that the ϕ-PDF tail is related to the deterministic portion of the dynamics while the emergence of the algebraic tail in the θ-PDF is due to the fact that optimal contribution, which defines the tail, is a long one, we indeed conclude that these results equally apply to both cases of thermal and velocity fluctuations. Analogously, the τ-PDF tail is exponential, regardless of whether or not the tumbling is driven by thermal or velocity fluctuations, because rare fluctuations producing the exponential tail can be interpreted in terms of a large number of unsuccessful attempts to pass the fluctuational domain.

We conclude with some general remarks related to the applicability conditions of our approach and the validity of the assumptions used in this letter. First, even though the major experimental motivation for this study is provided by existing and prospective elastic turbulence experiments, the situation when steady shear flow and weaker fluctuating velocity are super-imposed is actually a very common one. For example, the setting is typical for a boundary layer dynamics in moderate-to high Re-number turbulence in a pipe. Second, considering the statistics of tumbling driven by velocity fluctuations we restrict our attention here to discussing solely the state above the coil-stretch transition, when the Lyapunov exponent of the flow exceeds the polymer relaxation rate [9–11]. Third, this letter does not discuss the statistics of the polymer elongation which can be examined in the framework of a scheme similar to the one presented in this letter and which will become the subject of a separate publication in which a more detailed account for all calculations presented in the paper will also be given. Fourth, our considerations imply that the (Lagrange) velocity correlation time is less than or of the order of, the characteristic tumbling time τ_t. It is natural to expect that the velocity correlation time is equal to the inverse Lyapunov exponent λ^{-1}, which in the framework of our scheme is $\lambda^{-1} \sim \tau_t$, thus justifying the assumption. Fifth, typical parameters of flows in realistic experimental settings vary along the Lagrangian trajectory demonstrating essential spatial inhomogeneity. Even though these variations were not accounted for in our derivations, the results remain valid if the variations occur on time scale larger than τ_t. Then the PDFs discussed in the paper adjust adiabatically to the current values of the parameters and become, consequently, slow functions of a spatial position. Sixth, the description of this letter does not apply in the neighborhood of the walls, where the velocity modeling should be essentially modified [15]. Seventh, if the regular part of the flow is elongational the polymer flips becomes forbidden in the ideally deterministic regime while fluctuations will still generate some tumbling. An analysis of tumbling in the elongational flow is planned to be the subject of separate consideration. Finally, one should note that tumbling of vesicles [16, 17] is an interesting subject for future studies.

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